

FORM PTO-1390
(REV 10-94)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

10921.101USWO

U.S. APPLICATION NO. (if known, see 37 C.F.R. 1.5)

UNKNOWN 10/019810

INTERNATIONAL APPLICATION NO.

PCT/JP/02568

INTERNATIONAL FILING DATE

APRIL 19, 2000

PRIORITY DATE CLAIMED

APRIL 21, 1999

TITLE OF INVENTION

AQUEOUS POLYAMIDE RESIN DISPERSION AND PROCESS FOR PRODUCING THE SAME

APPLICANT(S) FOR DO/EO/US

ARAKI, Eiji; SUGIHARA, Norihiro; NAKAO, Kaichiro; MANABE, Hiroshi

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98, form 1449, and 8 references.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
☐ A SECOND of SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information: PCT/ISA/210; PCT/IB/304; PCT/IB/308; PCT/IB/332

U.S. APPLICATION NO. (If known, see 37 CFR 1.5) UNKNOWN		INTERNATIONAL APPLICATION NO. PCT/JP/02568		ATTORNEY'S DOCKET NUMBER 10921.101USWO	
---	--	---	--	---	--

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):

Search Report has been prepared by the EPO or JPO.....\$890.00

International preliminary examination fee paid to USPTO
(37 CFR 1.492(a)(1)).....\$710.00

No international preliminary examination fee paid to USPTO (37 CFR 1.482)
but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....\$740.00

Neither international preliminary examination fee (37 CFR 1.482) nor
international search fee (37 CFR 1.445(a)(3)) paid to USPTO \$1040.00

International preliminary examination fee paid to USPTO (37 CFR 1.482)
and all claims satisfied provisions of PCT Article 33(2)-(4).....\$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT = \$890.00

Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [] 30
months from the earliest claimed priority date (37 CFR 1.492(e)). **\$0**

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	18	-20 = 0	X \$18.00	\$0	
Independent claims	2	-3 = 0	X \$84.00	\$0	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ \$260.00	\$0
TOTAL OF ABOVE CALCULATIONS =				\$890.00	
Reduction by 1/2 for filing by small entity, if applicable. Small entity status is claimed pursuant to 37 CFR 1.27				\$0	
SUBTOTAL =				\$890.00	
Processing fee of \$130.00 for furnishing the English translation later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				+ \$0	
TOTAL NATIONAL FEE =				\$890.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+ \$40.00	
TOTAL FEES ENCLOSED =				\$930.00	
				Amount to be: refunded	\$0
				charged	\$0

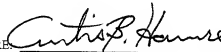
a. ☒ Check(s) in the amount of \$890.00 and \$40.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees.
A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 13-2725.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO
Douglas P. Mueller
MERCHANT & GOULD
P.O. Box 2903
Minneapolis, MN 55402-0903


SIGNATURE: 
NAME: Curtis B. Hamre
REGISTRATION NUMBER: 29,165

S/N unknown

PATENT #4/a

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Araki, et al.	Docket No.:	10921.101USWO
Serial No.:	unknown	Filed:	concurrent herewith
Int'l Appln No.:	PCTJP02568	Int'l Filing Date:	April 19, 2000
Title:	AQUEOUS POLYAMIDE RESIN DISPERSION AND PROCESS FOR PRODUCING THE SAME		

CERTIFICATE UNDER 37 CFR 1.10
 "Express Mail" mailing label number: EL669944235US
 Date of Deposit: October 19, 2001
 I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office To Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231.
 By: 
 Chris Stordahl

PRELIMINARY AMENDMENT

Box PCT
 Assistant Commissioner for Patents
 Washington, D. C. 20231

Dear Sir:

In connection with the above-identified application filed herewith, please enter the following preliminary amendment, a copy of which is enclosed herewith:

IN THE ABSTRACT

Insert the attached Abstract page into the application as the last page thereof.

IN THE SPECIFICATION

A courtesy copy of the present specification is enclosed herewith. However, the World Intellectual Property Office (WIPO) copy should be relied upon if it is already in the U.S. Patent Office.

1019810.101901

REMARKS

A new abstract page is supplied to conform to that appearing on the publication page of the WIPO application, but the new Abstract is typed on a separate page as required by U.S. practice.

Applicants respectfully request that the preliminary amendment described herein be entered into the record prior to calculation of the filing fee and prior to examination and consideration of the above-identified application.

If a telephone conference would be helpful in resolving any issues concerning this communication, please contact Applicants' primary attorney-of record, Douglas P. Mueller (Reg. No. 29,165), at (612) 371.5237.

Respectfully submitted,

MERCHANT & GOULD P.C.
P.O. Box 2903
Minneapolis, Minnesota 55402-0903
(612) 332-5300

Dated: October 19, 2001

By Curtis B. Hamre
Curtis B. Hamre
Reg. No. 29,165

CBH:DPM/rw



10/019810

ABSTRACT

The aqueous dispersion of polyamide resin of the present invention comprises dispersed polyamide resin particles, basic material and water. The dispersed polyamide resin particles have a weight-average diameter of 0.1-10 μm . The ratio of end carboxyl groups to end amino groups in the polyamide resin is between 60/40 and 100/0. The amount of said basic material added is 0.2-3.0 mol per mol of end carboxyl groups. Such an aqueous dispersion of polyamide resin can be manufactured by adding the polyamide resin to an aqueous dispersion medium containing 0.2-3.0 mol of basic material per mol of end carboxyl groups in the polyamide resin.

10019810-101901

CERTIFICATE UNDER 37 CFR 1.10

'Express Mail' mailing label number: EL669944235US

Date of Deposit: October 19, 2001

I hereby certify that this paper or fee is being deposited with the United States Postal Service 'Express Mail Post Office To Addressee' service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, D.C. 20231.

By:


Chris Stordahl

DESCRIPTION

AQUEOUS DISPERSION OF POLYAMIDE RESIN AND
METHOD OF MANUFACTURE THEREOF

5

TECHNICAL FIELD

The present invention relates to an aqueous dispersion of polyamide resin wherein polyamide resin particles are dispersed in an aqueous dispersion medium, and to a method of manufacture thereof.

10

BACKGROUND ART

Aqueous dispersions of polyamide resin can convey oil resistance, solvent resistance, chemical resistance, abrasion resistance and gas blocking and adhesive properties and the like when used as a basis in the formation of coating films. Consequently, aqueous dispersions of polyamide resin are widely used in water-color inks, textile treatments, textile fillers, paper treatments, binders, lubricants, steel plate finishing agents, surface modifiers, hot melt adhesives and the like.

15

20

It is difficult to prepare a dispersion of polyamide in an aqueous dispersion medium by a direct process of emulsion polymerization in the aqueous dispersion medium because of its manufacturing process. Therefore, various methods have been proposed for manufacturing aqueous dispersions of polyamide resin by dispersion in an aqueous medium of polyamide resin formed by condensation

25

polymerization or ring-opening polymerization. Possible methods of dispersing polyamide resin in an aqueous medium include reprecipitation and post-emulsification.

- Reprecipitation is a method of dissolving polyamide resin in an organic solvent, reprecipitating the polyamide resin and replacing the organic solvent with an aqueous medium (see for example Japanese Patent Applications Laid-open No. S61-223059 and S63-186738).

- The main problems with reprecipitation are the following. First, the polyamide resin particles obtained from reprecipitation are large in diameter, limiting the uses of the manufactured aqueous dispersion of polyamide resin. Second, polyamide resin particles often re-aggregate when the organic solvent is replaced with an aqueous medium, so the aqueous dispersion of polyamide resin obtained by reprecipitation is lacking in standing stability. Third, the inclusion of the step of replacing the organic solvent with an aqueous medium complicates the manufacturing process in the reprecipitation method.

- In the post-emulsification method polyamide resin is dispersed in an aqueous medium as follows. In the first step, polyamide resin is dissolved in a non-water-soluble or slightly water-soluble organic solvent to prepare a polyamide resin solution. In the second step, the polyamide resin solution is mixed together with an emulsifier in an aqueous medium to prepare a mixture. In the third step, the mixture is agitation emulsified at high shear force in a specialized emulsifying apparatus. In the fourth step, the

organic solvent is removed from the mixture, resulting in an aqueous dispersion of polyamide resin.

The main problems with post-emulsification are the following. First, because polyamide resin is not very soluble with respect to organic solvents, the post-emulsion method has low productivity and cannot be called economical. Second, because bubbling occurs during removal of the organic solvent in the post-emulsification method, a process is required to control bubbling, complicating the operation and making the method less economical. Third, the aqueous dispersion of polyamide resin obtained by post-emulsification inevitably contains residues of organic solvent and emulsifiers. Fourth, the post-emulsification method uses large quantities of organic solvent which could contaminate the work environment and cause environmental pollution.

DISCLOSURE OF THE INVENTION

It is an object of the present invention to resolve the various issues discussed above.

The aqueous dispersion of polyamide resin provided by the first aspect of the present invention contains dispersed polyamide resin particles, basic material and water, with the dispersed polyamide resin particles having a weight-average diameter of 0.1-10 μm and a ratio of end carboxyl groups to end amino groups being between 60/40 and 100/0, and with the amount of said basic material being 0.2-3.0 mol per mol of said end carboxyl groups.

Polyamide resin has carboxyl and/or amino groups on the ends. In general, carboxyl groups have a relatively low degree of electrolytic dissociation with respect to water. However, coexisting carboxyl groups with basic material
5 promotes electrolytic dissociation of carboxyl groups in water solution (aqueous dispersion). Consequently, addition of basic material to the aforementioned aqueous dispersion of polyamide resin promotes electrolytic dissociation of end carboxyl groups on the dispersed
10 polyamide resin particles, and improves dispersive power with respect to an aqueous dispersion medium.

The cause of aggregation of dispersed polyamide resin particles is attractive force acting on the particles. One such attractive force can be attributed to hydrogen bond.

15 The aforementioned dispersed polyamide resin particles have relatively few amino groups in proportion to carboxyl groups. Consequently, with the aforementioned aqueous dispersion of polyamide resin there is relatively little hydrogen bonding force acting among the dispersed polyamide resin
20 particles, and excessive aggregation of the dispersed polyamide resin particles is therefore prevented.

Thus, because excessive aggregation of the dispersed polyamide resin particles is controlled with the aforementioned aqueous dispersion of polyamide resin,
25 standing stability is excellent.

An alkali metal hydroxide or amino compound is preferred as the basic material.

Examples of possible alkali metal hydroxide include sodium hydroxide and potassium hydroxide.

In the present invention, the term "amino compound" also includes ammonia.

- 5 The method of manufacturing an aqueous dispersion of polyamide resin provided by the second aspect of the present invention is characterized by the addition of polyamide resin to an aqueous dispersion medium containing 0.2-3.0 mol of basic material per mol of end carboxyl groups on the
- 10 polyamide resin.

As mentioned already, electrolytic dissociation of carboxyl groups is promoted by the coexistence of basic material with polyamide resin, allowing for the prevention of excessive aggregation between polyamide resin particles.

- 15 The aforementioned method of manufacturing an aqueous dispersion of polyamide resin employs an aqueous dispersion medium containing basic material, providing an aqueous dispersion of polyamide resin with small-diameter polyamide resin particles and excellent standing stability.

- 20 The amount of said basic material in the aforementioned aqueous dispersion medium should be in the range of 0.2-3.0 mol per mol of said end carboxyl groups. If the amount of said basic material falls below this range, aggregation of polyamide resin particles will not be sufficiently
- 25 controlled, making it difficult to obtain an aqueous dispersion of polyamide resin. If the amount of said basic material exceeds this range, the resulting aqueous

dispersion of polyamide resin will be impractical due to high alkalinity.

Preferably, the amount of said basic material should be 0.4-2.0 mol per mol of said end carboxyl groups.

- 5 Particularly favorable results are obtained if the amount of said basic material is 0.6-1.5 mol.

The ratio of said end carboxyl groups to said end amino groups in the polyamide resin should be between 60/40 and 100/0.

- 10 As mentioned above, amino groups are one cause of aggregation of polyamide resin particles because of their susceptibility to hydrogen bonding. The polyamide resin used in the aforementioned method of manufacturing an aqueous dispersion of polyamide resin has a relatively small number of end amino groups in proportion to end carboxyl groups.
- 15 Consequently, the aforementioned manufacturing method avoids excessive aggregation caused by end amino groups, and provides an aqueous dispersion of polyamide resin with excellent standing stability.

- 20 The polyamide resin used in the aforementioned method of manufacturing an aqueous dispersion of polyamide resin should have 50-3000 mmol of end carboxyl groups per kg of said polyamide resin. If the amount of end carboxyl groups falls below this range, it will be impossible to adequately
- 25 disperse the polyamide resin in the aqueous dispersion medium. The upper limit is set at 3000 mmol because there are limits on the amount of end carboxyl groups that can be introduced into polyamide resin. However, because the

dispersive force of polyamide resin in aqueous dispersion medium increases as the amount of end carboxyl groups in the polyamide resin increases, the amount of end carboxyl groups should be as large as is economically feasible.

- 5 In order to obtain even better results, the amount of end carboxyl groups in the polyamide resin is in the range of 100-2000 mmol per kg of polyamide resin.

- In the method of manufacturing an aqueous dispersion of polyamide resin of the present invention, a polyamide
10 resin containing as a structural unit at least one from the group of $-\text{[NH(CH}_2\text{)}_5\text{CO]}-$, $-\text{[NH(CH}_2\text{)}_6\text{NHCO(CH}_2\text{)}_4\text{CO]}-$, $-\text{[NH(CH}_2\text{)}_5\text{NHCO(CH}_2\text{)}_8\text{CO]}-$, $-\text{[NH(CH}_2\text{)}_{10}\text{CO]}-$ and $-\text{[NH(CH}_2\text{)}_{11}\text{CO]}$ is used by preference.

- Specific examples of polyamide resin include 6-nylon,
15 66-nylon, 610-nylon, 11-nylon, 12-nylon, 6/66 copolymer nylon, 6/610 copolymer nylon, 6/11 copolymer nylon, 6/12 copolymer nylon, 6/66/11 copolymer nylon, 6/66/12 copolymer nylon, 6/66/11/12 copolymer nylon and 6/66/610/11/12 copolymer nylon. In the said method of manufacturing an
20 aqueous dispersion of polyamide resin, the listed polymers or copolymers can be used individually or more than one can be used in combination as the polyamide resin.

- The polyamide resin used in the said method of manufacturing an aqueous dispersion of polyamide resin can
25 be manufactured by a well-known method. For example, methods that can be used for manufacturing the polyamide resin include condensation polymerization of diamines with dicarboxylic acids, condensation polymerization of ω-

amino- ω '-carboxylic acids, or ring-opening polymerization of ring lactams.

- Specific examples of diamines include ethylene diamine, trimethylene diamine, tetramethylene diamine, 5 pentamethylene diamine, hexamethylene diamine, 1,7-diaminoheptane, 1,8-diaminooctane, 1,9-diaminononane, 1,10-diaminodecane, phenylene diamine, and methaxylilene diamine.

- Specific examples of dicarboxylic acids include 10 glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, nonane dicarboxylic acid, decane dicarboxylic acid, tetradecane dicarboxylic acid, octadecane dicarboxylic acid, fumaric acid, phthalic acid, and xylilene dicarboxylic acid.

- 15 Specific examples of ω -amino- ω '-carboxylic acids include 6-aminocaproic acid, 7-aminoheptanoic acid, 9-aminononanoic acid, 11-aminoundecanoic acid, and 12-aminododecanoic acid.

- Specific examples of ring lactams include ϵ -caprolactam, 20 ω -enantholactam and ω -lauryllactam.

- In order to adjust the proportion of end carboxyl groups to end amino groups in the polyamide resin to between 60/40 and 100/0, a specific amount of a dicarboxylic or monocarboxylic acid can be added as a polymerization 25 regulator during condensation polymerization or ring-opening polymerization.

Specific examples of dicarboxylic acids that can be used as the said polymerization regulator include glutaric acid,

adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, nonane dicarboxylic acid, decane dicarboxylic acid, tetradecane dicarboxylic acid, octadecane dicarboxylic acid, fumaric acid, phthalic acid and xylilene
5 dicarboxylic acid.

Specific examples of monocarboxylic acids that can be used as the said polymerization regulator include caproic acid, heptanoic acid, nonanoic acid, undecanoic acid and dodecanoic acid.

- 10 Possible basic materials for mixing with the aqueous dispersion medium include alkali metal hydroxides or amino compounds.

Examples of said alkali metal hydroxides include lithium hydroxide, sodium hydroxide, potassium hydroxide,
15 rubidium hydroxide, cesium hydroxide and francium hydroxide. Of these alkali metal hydroxides, sodium hydroxide and potassium hydroxide are used by preference in the present invention since they effectively enhance the dispersive force of the polyamide resin.

- 20 Well-known amino compounds including ammonia can be used as the said amino compounds.

The amount of water in the aqueous dispersion medium should be 30-1500 parts by weight based on 100 parts by weight of polyamide resin. If the amount of water falls below this
25 range, the polyamide resin cannot be adequately dispersed in the aqueous dispersion medium, while if the amount of water exceeds this range, the concentration of polyamide

resin in the resulting aqueous dispersion of polyamide resin will be excessively low.

- 5 In order to obtain still better results, the amount of water is kept in the range of 100-500 parts by weight based on 100 parts by weight of polyamide resin.

- 10 A variety of additives including high polymer dispersants, inorganic dispersants, anionic surfactants, nonionic surfactants, ampholytic surfactants and antioxidants may be added to the aqueous dispersion medium as long as they do not detract from the properties of the resulting aqueous dispersion of polyamide resin.

Examples of high polymer dispersants include polyacrylate, polystyrene sulfonate, styrene anhydrous maleate, polyvinyl alcohol and hydroxyethyl cellulose.

- 15 Examples of inorganic dispersants include alumina sol, silica sol and calcium phosphate.

Examples of anionic surfactants include rosin acid salts, fatty acid salts and alkylbenzene sulfonate.

- 20 Examples of nonionic surfactants include polyoxyethylene alkyl ether, glycerine fatty acid ester and polyoxyethylene fatty acid ethanolamide.

- 25 In the preferred embodiments, the polyamide resin is heated to a temperature above the softening point of the polyamide resin for dispersion in the aqueous dispersion medium. Heating the polyamide resin to above this softening point releases the associations between molecules and aggregation among particles. Consequently, the diameter of the polyamide resin particles can be minimized. Also, as

10019810-101901

explained above, aggregation (or re-aggregation) among polyamide resin particles is controlled by the addition of basic material and by the presence of carboxyl groups, so the particles of polyamide resin are maintained in the aqueous dispersion with a small diameter in a suitable dispersed state.

The heating temperature of the polyamide resin cannot be stated univocally because it is determined for example according to the melting point of the polyamide resin (the type and degree of polymerization of the structural units). However, if the heating temperature is too low, the polyamide does not soften sufficiently and cannot be dispersed in the aqueous dispersion medium, while if the heating temperature is too high the polyamide resin is vulnerable to degradation. Consequently, the heating temperature of the polyamide resin is generally in the range of 70-300°C or preferably 90-220°C.

Ideally, the polyamide resin is dispersed in the aqueous dispersion medium by application of shear force to an aqueous dispersion medium to which the polyamide resin has been added.

When applied to an aqueous dispersion medium to which polyamide resin has been added, shear force releases associations between molecules and/or aggregation among particles of the polyamide resin. This results not only in small diameter polyamide resin particles, but also in uniform dispersion of the polyamide resin particles in the aqueous dispersion medium.

- Application of shear force to the aqueous dispersion medium can be achieved for example by agitation of the aqueous dispersion medium. The aqueous dispersion medium can be agitated for example by rotation of mixing blades.
- 5 In this case, the rotational frequency of the mixing blades is set for example at 100-500 times a minute. The rotational frequency of the mixing blades is set above 100 times a minute in order to obtain an adequate agitation effect. Rotation of the mixing blades over 500 times a minute provides no
- 10 benefit proportional to the increased rotation and only increases costs.

BEST MODE FOR CARRYING OUT THE INVENTION

- The following are two possible means of manufacturing
- 15 the aqueous dispersion of polyamide resin of the present invention.

The first manufacturing method comprises the following steps.

- In the first step, specific quantities of polyamide
- 20 resin, water and basic material are supplied in one batch to a dispersion tank.

- In the second step, the aqueous dispersion medium is agitated using a mixing blade at 100-500 rpm while being heated to a temperature above the softening temperature of
- 25 the polyamide resin in the aqueous dispersion medium, raising the temperature of both aqueous dispersion medium and polyamide resin.

10019810-101901

In the third step, the temperature of both aqueous dispersion medium and polyamide resin is maintained at a temperature above the softening temperature of the polyamide resin, while agitation is continued for about 10-60 minutes using a mixing blade at 100-500 rpm.

The second manufacturing method comprises the following steps.

10 In the first step, a dispersion tank is first heated to a temperature above the softening temperature of the polyamide resin in the aqueous dispersion medium, and kept under pressure.

15 In the second step, the heating and pressure conditions of the dispersion tank are maintained, and fixed amounts of melted polyamide resin, water and basic material are added separately one after another to the dispersion tank with a mixing blade being rotated at 100-500 rpm.

20 In the third step, agitation is continued for about 10-60 minutes using a mixing blade at 100-500 rpm while maintaining the dispersion tank at a temperature above the softening temperature of the polyamide resin in the aqueous dispersion medium.

25 The dispersion tank used in the aforementioned methods may be any pressure proof container equipped with a means for heating to a temperature above the softening temperature of the polyamide resin in the aqueous dispersion medium and a means of agitation sufficient to apply shear force to the contents. For example, a pressure proof autoclave equipped

with an agitator can be used as the dispersion tank in the
aforementioned manufacturing methods.

- In either one of the aforementioned manufacturing
methods, the polyamide resin is subject to shear force from
5 agitation while in a softened state in the aqueous dispersion
medium. Consequently, the associations between molecules
and aggregations of particles of the polyamide resin are
released, the particle diameter is minimized, and dispersal
is uniform in the aqueous dispersion medium. Moreover,
10 electrolytic dissociation of end carboxyl groups in the
polyamide resin is promoted by the effect of the basic
material. The disassociated carboxyl groups (carboxylate
ions) serve the purpose of a stabilized emulsifier in the
aqueous dispersion medium, enhancing the hydrophilicity of
15 the polyamide resin as a whole and preventing excessive
aggregation between molecules in the manufacturing process.
As a result, either of the aforementioned manufacturing
methods results in dispersion in an aqueous dispersion
medium of fine polyamide particles with a weight average
20 particle diameter of 0.1-10 μ m.

The aqueous dispersion of polyamide resin obtained in
this way can be adjusted to the desired concentration using
a suitable concentration means such as a semipermeable
membrane.

- 25 It is also possible to use the resulting aqueous
dispersion of polyamide resin in the form of a fine powder
obtained by a drying method such as spray drying, either

as is or following solid-liquid separation by centrifugation or filtration.

- Next, the present invention is explained in more detail using examples and comparative examples. However, the scope of the technical concept of the present invention is not limited by these examples.

[Example 1]

- In this example, the raw materials were placed in an autoclave equipped with a turbine type agitator and a heating jacket, the autoclave was sealed, and an aqueous dispersion of polyamide resin was manufactured by heating the raw materials while applying shear force.

- The autoclave had an internal diameter of 700 mm, a height of 1500 mm and a content volume of 450 L. The mixing blade had a diameter of 350 mm. The heating jacket worked by the circulation of heated oil.

- The raw materials were 120 kg of 6/66/12 copolymer nylon, 179.6 kg of water and 0.4 kg (10 mol) of sodium hydroxide.
- In the 6/66/12 copolymer nylon, the ratio of end carboxyl groups to end amino groups was 87/13, and the amount of end carboxyl groups was 130 mmol per kg of polyamide resin.

- The autoclave was heated to 150°C and then maintained at 150°C for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

After being heated and mixed in this way, the contents of the autoclave were cooled to 50°C, and removed from the

autoclave as the aqueous dispersion of polyamide resin of the present invention.

In this aqueous dispersion of polyamide resin, the weight average particle diameter of the polyamide resin was 1.2 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000). Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

[Example 2]

In this example, an aqueous dispersion of polyamide resin was manufactured under the same conditions as in Example 1, except that the raw materials were 6/66/12 copolymer nylon having a ratio of end carboxyl groups to end amino groups of 92/8 and an amount of end carboxyl groups of 165 mmol per kg of polyamide resin, together with 178.8 kg of water and 1.2 kg (21.8 mol) of potassium hydroxide.

In this aqueous dispersion of polyamide resin, the weight average particle diameter of the polyamide resin was 0.3 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000). Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

[Example 3]

In this example, the raw materials were 120 kg of 6/66/11/12 copolymer nylon, 179.2 kg of water and 0.8 kg (20.0 mol) of sodium hydroxide. The 6/66/11/12 copolymer nylon had a ratio of end carboxyl groups to end amino groups of 72/28 and an amount of end carboxyl groups of 170 mmol per kg of polyamide resin. Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 170°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

In the resulting aqueous dispersion of polyamide resin, the weight average particle diameter of the polyamide resin was 1.1 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000). Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

[Example 4]

In this example, the raw materials were 120 kg of 6/12 copolymer nylon, 179.4 kg of water and 0.6 kg (10.7 mol) of potassium hydroxide. The 6/12 copolymer nylon had a ratio of end carboxyl groups to end amino groups of 88/12 and an amount of end carboxyl groups of 120 mmol per kg of polyamide resin. Using these raw materials and an autoclave similar

to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 170°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was

5 150 times a minute.

In the resulting aqueous dispersion of polyamide resin, the weight average particle diameter of the polyamide resin was 1.8 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000).

10 Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

15 [Example 5]

In this example, the raw materials were 120 kg of 6/66/610/11/12 copolymer nylon, 179.5 kg of water and 0.5 kg (12.5 mol) of sodium hydroxide. The 6/66/610/11/12 copolymer nylon had a ratio of end carboxyl groups to end amino groups of 66/34 and an amount of end carboxyl groups of 120 mmol per kg of polyamide resin. Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 150°C and then
20 maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

In the resulting aqueous dispersion of polyamide resin, the weight average particle diameter of the polyamide resin

was 2.3 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000). Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent
5 standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

[Example 6]

In this example, the raw materials were 120 kg of
10 12-nylon, 179.2 kg of water and 0.4 kg (10.0 mol) of sodium hydroxide. The 12-nylon had a ratio of end carboxyl groups to end amino groups of 85/15 and an amount of end carboxyl groups of 90 mmol per kg of polyamide resin. Using these raw materials and an autoclave similar to that used in
15 Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 200°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

In the resulting aqueous dispersion of polyamide resin,
20 the weight average particle diameter of the polyamide resin was 3.8 μm as measured with a laser diffraction particle size distribution meter (Shimadzu Seisakusho SALD 2000). Observation of the aqueous dispersion of polyamide resin after it had been left for 1 month at 50°C showed excellent
25 standing stability, with absolutely no aggregative or floating disassociation of the polyamide resin.

[Comparative Example 1]

In this comparative example, the raw materials were 120 kg of 6/66/12 copolymer nylon and 180.0 kg of water. In other words, no basic compound was included in the raw materials of this comparative example. The 6/66/12 copolymer nylon was similar to that used in Example 1.

Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 150°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

The contents obtained in this way were a lump of polyamide resin rather than an aqueous dispersion.

[Comparative Example 2]

In this comparative example, the raw materials were 120 kg of 6/66/12 copolymer nylon, 179.6 kg of water and 0.1 kg (2.5 mol) of sodium hydroxide. The 6/66/12 copolymer nylon was similar to that used in Example 1.

Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was heated to 150°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

The contents obtained in this way were in the form of aggregated polyamide resin, not an aqueous dispersion.

[Comparative Example 3]

In this comparative example, the raw materials were 120 kg of 6/66/12 copolymer nylon, 179.6 kg of water and 0.14 kg (3.5 mol) of sodium hydroxide. The 6/66/12 copolymer nylon
5 had a ratio of end carboxyl groups to end amino groups of 18/82, and an amount of end carboxyl groups of 30 mmol per kg of polyamide resin.

Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide
10 resin was manufactured. The inside of the autoclave was heated to 150°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

The contents obtained in this way were in the form of
15 aggregated polyamide resin, not an aqueous dispersion.

[Comparative Example 4]

In this comparative example, the raw materials were 120 kg of 6/66/12 copolymer nylon, 179.6 kg of water and 0.4
20 kg (10.0 mol) of sodium hydroxide. The 6/66/12 copolymer nylon was similar to that used in Comparative Example 3.

Using these raw materials and an autoclave similar to that used in Example 1, an aqueous dispersion of polyamide resin was manufactured. The inside of the autoclave was
25 heated to 150°C and then maintained at that temperature for 30 minutes. The rotational speed of the mixing blade was 150 times a minute.

The contents obtained in this way were in the form of aggregated polyamide resin, not an aqueous dispersion.

10019810.101901

CLAIMS

1. An aqueous dispersion of polyamide resin, comprising:
dispersed polyamide resin particles, basic material
5 and water; and
wherein the weight average diameter of said dispersed
polyamide resin particles is 0.1-10 μm ,
the ratio of end carboxyl groups to end amino groups
in said polyamide resin is between 60/40 and 100/0, and
10 the amount of said basic material added is 0.2-3.0 mol
per mol of said end carboxyl groups.
2. The aqueous dispersion of polyamide resin according to
Claim 1, wherein said basic material is an alkali metal
15 hydroxide or amino compound.
3. The aqueous dispersion of polyamide resin according to
Claim 2, wherein said alkali metal hydroxide is sodium
hydroxide or potassium hydroxide.
- 20 4. The aqueous dispersion of polyamide resin according to
Claim 1, wherein the amount of said end carboxyl groups is
50-3000 mmol per kg of said polyamide resin.
- 25 5. The aqueous dispersion of polyamide resin according to
Claim 1, wherein said polyamide resin has as a structural
unit at least one selected from the group consisting of

-[NH(CH₂)₅CO]-, -[NH(CH₂)₆NHCO(CH₂)₄CO]-, -
NH(CH₂)₆NHCO(CH₂)₈CO]-, -[NH(CH₂)₁₀CO]- and -[NH(CH₂)₁₁CO].

6. The aqueous dispersion of polyamide resin according to
5 Claim 1, wherein the proportion of said water is 30-1500
parts by weight based on 100 parts by weight of polyamide
resin.

7. A method of manufacturing an aqueous dispersion of
10 polyamide resin, wherein polyamide resin is added to an
aqueous dispersion medium containing 0.2-3.0 mol of basic
material per mol of end carboxyl groups in said polyamide
resin.

- 15 8. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 7, wherein the ratio of
end carboxyl groups to end amino groups in said polyamide
resin is between 60/40 and 100/0.

- 20 9. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 7, wherein the amount
of said end carboxyl groups is 50-3000 mmol per kg of said
polyamide resin.

- 25 10. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 7, wherein said polyamide
resin has as a structural unit at least one selected from
the group consisting of -[NH(CH₂)₅CO]-, -

$[\text{NH}(\text{CH}_2)_6\text{NHCO}(\text{CH}_2)_4\text{CO}]_n-$, $-\text{[NH}(\text{CH}_2)_6\text{NHCO}(\text{CH}_2)_8\text{CO}]_n-$, $-\text{[NH}(\text{CH}_2)_{10}\text{CO}]_n-$ and $-\text{[NH}(\text{CH}_2)_{11}\text{CO}]_n-$.

11. The method of manufacturing an aqueous dispersion of
5 polyamide resin according to Claims 7, wherein said basic material is an alkali metal hydroxide or amino compound.

12. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 11, wherein said alkali
10 metal hydroxide is sodium hydroxide or potassium hydroxide.

13. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claims 7, wherein said aqueous
dispersion medium contains 30-1500 parts by weight of water
15 based on 100 parts by weight of said polyamide resin.

14. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 7, wherein said polyamide
resin is dispersed in said aqueous dispersion medium in a
20 state where the polyamide resin is heated to a temperature at or above the softening temperature of the resin.

15. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 14, wherein said
25 polyamide resin is heated at a temperature of 70°C-300°C.

16. The method of manufacturing an aqueous dispersion of
polyamide resin according to Claim 15, wherein said

polyamide resin is dispersed in said aqueous dispersion medium with shear force applied to the aqueous dispersion medium to which the polyamide resin has been added.

- 5 17. The method of manufacturing an aqueous dispersion of polyamide resin according to Claim 16, wherein shear force is applied to said aqueous dispersion medium by rotation of a mixing blade.

- 10 18. The method of manufacturing an aqueous dispersion of polyamide resin according to Claims 17, wherein the rotational speed of said mixing blade is 100-500 rpm.

10019810-101901

Declaration and Power of Attorney For Patent Application

特許出願宣言書

Japanese Language Declaration

私は、下欄に氏名を記載した発明者として、以下のとおり宣言する：

As a below named inventor, I hereby declare that:

私の住所、郵便の宛先および国籍は、下欄に氏名に続いて記載したとおりであり、

My residence, post office address and citizenship are as stated below next to my name,

名称の発明に関し、請求の範囲に記載した特許を求める主題の本来の、最初にして唯一の発明者である（一人の氏名のみが下欄に記載されている場合）か、もしくは本来の、最初にして共同の発明者である（複数の氏名が下欄に記載されている場合）と信じ、

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

AQUEOUS DISPERSION OF

POLYAMIDE RESIN AND

METHOD OF MANUFACTURE THEREOF

その明細書を
(該当する方に印を付す)

the specification of which
(check one)

☐ここに添付する。

☐Is attached hereto.

☐ _____ 日に出席番号
第 _____ 号として提出し、
_____ 日に補正した。
(該当する場合)

☐ was filed on _____ as
Application Serial No. _____
and was amended on _____
(if applicable)

☐ _____ 日にPCT国際出願番号
第 _____ 号として提出し、
PCT第19条に基づき _____ 日に補正した。
(該当する場合)

☒ was described and claimed in PCT international
application No. PCT/JP00/02568
filed on APRIL 19, 2000
and as amended under PCT Article 19 or 34 on _____
(if applicable)

私は、前記のとおり補正した請求の範囲を含む前記明細書の内容を検討し、理解したことを陳述する。

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

私は、連邦規則法典第37部第1章第56条(a)項に従い、本願の審査に所要の情報を開示すべき義務を有することを認める。

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56(a).

Japanese Language Declaration

私は、合衆国法典第35部第119条 (a) - (d) 項または第365条 (a) - (b) 項にもとづく下記の外国特許出願または発明者証出願または少なくとも1つの合衆国以外の国を指定した PCT 国際出願の外国優先権利益を主張し、さらに優先権の主張に係る基礎出願の出願日前の出願日を有する外国特許出願または発明者証出願または PCT 国際出願を以下に明記する:

I hereby claim foreign priority benefits under Title 35, United States Code, § 119(a)-(d) or § 365(a)-(b) of any foreign application(s) for patent or inventor's certificate, or of any PCT international application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate or PCT international application having a filing date before that of the application on which priority is claimed:

Prior foreign applications

先の外国出願

(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願の年月日)
Patent Application No. 11-113056	Japan	21/4/1999
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願の年月日)
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願の年月日)
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願の年月日)
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願の年月日)

Priority claimed

優先権の主張

<input checked="" type="checkbox"/> Yes あり	<input type="checkbox"/> No なし
<input type="checkbox"/> Yes あり	<input type="checkbox"/> No なし
<input type="checkbox"/> Yes あり	<input type="checkbox"/> No なし
<input type="checkbox"/> Yes あり	<input type="checkbox"/> No なし
<input type="checkbox"/> Yes あり	<input type="checkbox"/> No なし

私は、合衆国法典第35部第120条にもとづく下記の合衆国特許出願の利益または第365条 (c) 項にもとづく合衆国を指定する PCT 国際出願の利益を主張し、本願の請求の範囲各項に記載の主題が合衆国法典第35部第112条第1項に規定の態様で先の合衆国出願に開示されていない限度において、先の出願の出願日と本願の国内出願日または PCT 国際出願日の間に公表された連邦規則法典第37部第1章第56条 (a) 項に記載の所要の情報を開示すべき義務を有することを認める:

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT international application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, § 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.) (出願番号)	(Filing Date) (出願日)	(状況) (特許済み、係属中、放棄済み)	(Status) (patented, pending, abandoned)
(Application Serial No.) (出願番号)	(Filing Date) (出願日)	(状況) (特許済み、係属中、放棄済み)	(Status) (patented, pending, abandoned)

Japanese Language Declaration

私は、ここに自己の知識にもとづいて行った陳述がすべて真実であり、自己の有所する情報および信ずるところに従って行った陳述が真実であると信じ、さらに故意に虚偽の陳述を行った場合、合衆国法典第18部第1001条により、罰金もしくは禁固に処せられるか、またはこれらの刑が併科され、またかかる故意による虚偽の陳述が本願ないし本願に対して付与される特許の有効性を損なうことがあることを認識して、以上の陳述を行ったことを宣言する。

委任状：私は、下記発明者として、以下の代理人をここに選任し、本願の手続を遂行すること並びにこれに関する一切の行為を特許商標庁に対して行うことを委任する。(代理人氏名および登録番号を明記のこと)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (list name and registration number)

Bartingale, Karl H. Reg. No. 35,183
Batzli, Brian H. Reg. No. 32,960
Beck, Robert C. Reg. No. 28,184
Bogucki, Raymond A. Reg. No. 17,425
Brennan, Thomas F. Reg. No. 35,075
Bruss, Steven C. Reg. No. 34,130
Byrne, Linda M. Reg. No. 32,404
Carlson, Alan G. Reg. No. 25,959
Caspers, Philip P. Reg. No. 33,227
Clifford, John A. Reg. No. 30,247
Conrad, Timothy R. Reg. No. 30,164
DiPietro, Mark J. Reg. No. 22,707
Edell, Robert T. Reg. No. 20,187
Freed, Robert C. Reg. No. 32,569
Gates, George H. Reg. No. 33,500
Golla, Charles E. Reg. No. 28,895
Gould, John D. Reg. No. 18,223
Gresens, John J. Reg. No. 33,172
Hamre, Curtis B. Reg. No. 23,155
Hassing, Thomas A. Reg. No. 35,159
Hillson, Randall A. Reg. No. 31,838

Johnson, David G. Reg. No. 31,628
Khuth, Daniel J. Reg. No. 32,146
Kowalchuk, Alan W. Reg. No. 31,535
Lasky, Michael B. Reg. No. 29,555
Lundberg, Steven W. Reg. No. 30,568
Lynch, David W. Reg. No. 36,204
Mau, Michael L. Reg. No. 30,087
Maxin, John L. Reg. No. 34,668
McDonald, Daniel W. Reg. No. 32,044
McDonald, Wendy M. Reg. No. 32,427
Michel, Michelle M. Reg. No. 33,968
Moy, R. Carl Reg. No. 30,725
Mueeting, Ann M. Reg. No. 33,977
Mundelius, Antony C. Reg. No. 35,963
Nelson, Albin J. Reg. No. 28,650
Raasch, Kevin W. Reg. No. 35,651
Retland, Earl D. Reg. No. 25,767
Rothfus, Joel A. Reg. No. 33,277
Schmidt, Cecil C. Reg. No. 20,566
Schuman, Mark D. Reg. No. 31,197
Schumann, Michael D. Reg. No. 30,422

Schwappach, Karl G. Reg. No. 35,786
Schwegman, Micheal L. Reg. No. 25,816
Sebald, Gregory A. Reg. No. 33,280
Smith, Phillip H. Reg. No. 20,476
Smith, Stephanie J. Reg. No. 34,437
Sorensen, Andrew D. Reg. No. 33,606
Stenstrom, Dennis G. Reg. No. 28,910
Strawbridge, Douglas A. Reg. No. 28,376
Strodthoff, Kristine M. Reg. No. 34,259
Sumner, John P. Reg. No. 29,114
Sumners, John S. Reg. No. 24,216
Taylor, Gregory M. Reg. No. 34,263
Telleson, David K. Reg. No. 32,314
Underhill, Albert L. Reg. No. 27,403
Vandenburgh, J. Derek Reg. No. 32,179
Welter, Paul A. Reg. No. 20,890
Williams, Douglas J. Reg. No. 27,054
Woessner, Warren D. Reg. No. 30,440
Wood, Gregory B. Reg. No. 28,133

書類の送付先：
Douglas P. Mueller
MERCHANT & GOULD P.C.
3200 IDS Center, 80 South 8th Street,
Minneapolis, MN 55402-2215, U.S.A.

Send Correspondence to:
Douglas P. Mueller
MERCHANT & GOULD P.C.
3200 IDS Center, 80 South 8th Street,
Minneapolis, MN 55402-2215, U.S.A.

直通電話連絡先：(電話番号)
Telephone: 612/371-5237

Direct Telephone Calls to: (telephone number)
Telephone: 612/371-5237

Japanese Language Declaration

唯一のまたは第一の発明者の氏名	1-00	Full name of sole or first inventor <u>Eiichi Araki</u>
同発明者の署名	日付	Inventor's signature Date <u>Eiichi Araki</u> October 12, 2001
住所		Residence c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory Hyogo, Japan JPX
国籍		Citizenship Japan
郵便の宛先		Post Office Address c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory 1, Irifune-cho, Shikama-ku, Himeji-shi, Hyogo 672-8076 Japan

第2の共同発明者の氏名 (該当する場合)	2-00	Full name of second joint inventor, if any <u>Norihiro Sugihara</u>
同第2発明者の署名	日付	Second Inventor's signature Date <u>Norihiro Sugihara</u> October 12, 2001
住所		Residence c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory Hyogo, Japan JPX
国籍		Citizenship Japan
郵便の宛先		Post Office Address c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory 1, Irifune-cho, Shikama-ku, Himeji-shi, Hyogo 672-8076 Japan

第3の共同発明者の氏名 (該当する場合)	3-00	Full name of third joint inventor, if any <u>Kaichiro Nakao</u>
同第3発明者の署名	日付	Third Inventor's signature Date <u>Kaichiro Nakao</u> October 12, 2001
住所		Residence c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory Hyogo, Japan JPX
国籍		Citizenship Japan
郵便の宛先		Post Office Address c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory 1, Irifune-cho, Shikama-ku, Himeji-shi, Hyogo 672-8076 Japan

第4の共同発明者の氏名 (該当する場合)	4-00	Full name of fourth joint inventor, if any <u>Hiroshi Manabe</u>
同第4発明者の署名	日付	Fourth Inventor's signature Date <u>Hiroshi Manabe</u> October 12, 2001
住所		Residence c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory Hyogo, Japan JPX
国籍		Citizenship Japan
郵便の宛先		Post Office Address c/o Sumitomo Seika Chemicals Co., Ltd. Functional Polymers Research Laboratory 1, Irifune-cho, Shikama-ku, Himeji-shi, Hyogo 672-8076 Japan

第5の共同発明者の氏名 (該当する場合)		Full name of fifth joint inventor, if any
同第5発明者の署名	日付	Fifth Inventor's signature Date
住所		Residence
国籍		Citizenship
郵便の宛先		Post Office Address